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(54) Title: DUCTILE AND SOLVENT RESISTANT POLYCARBONATE COMPOSITIONS HAVING IMPROVED FLA- ME RESISTANCE		
(57) Abstract		
Ductile and solvent resistant aromatic polycarbonate compositions are obtained comprising an admixture of a high molecular weight aromatic polycarbonate and a block copolymer consisting of alternating segments of an aromatic polycarbonate and a polyorganosiloxane. The polycarbonate compositions also exhibit improved flame retardance.		

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Description

Ductile and Solvent Resistant Polycarbonate
Compositions Having Improved Flame Retardance

This invention relates to ductile and solvent resistant aromatic polycarbonate compositions that also have improved flame retardance.

Background of the Invention

Polycarbonate polymers are known as being excellent molding materials since products made therefrom exhibit such properties as high impact strength, toughness, high transparency, wide temperature limits (high impact resistance below -60°C and a UL thermal endurance rating of 115°C with impact), good dimensional stability, good creep resistance, good flame retardance, and the like. It would be desirable to add to this list of properties those of ductility and solvent resistance enabling these polycarbonate compositions to be employed to form molded articles that can be used in such applications as aircraft tray tables and seat backs, aircraft ducting, ski boots, and the like wherein the articles will be required to exhibit high tensile properties and resistance to the corrosive effects of commercial cleaning compounds and other organic chemicals.

Summary of the Invention

It has now been found that ductility and solvent resistance as well as improved flame retardance can be imparted to high molecular weight, aromatic polycarbonate resins by mixing the polycarbonate resin with block copolymers consisting of alternating segments of polybisphenol carbonates and polyorganosiloxane in amounts of about 1-30% by weight, preferably about 4-10% by weight, of the polycarbonate resin.



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In the practice of this invention, any of the aromatic polycarbonates can be employed that are prepared by reacting a diphenol with a carbonate precursor. Typical of some of the diphenols that can be employed are bisphenol-A (2,2-bis(4-hydroxy-phenyl)propane), bis(4-hydroxyphenyl)methane, 2,2-bis(4-hydroxy-3-methylphenyl)propane, 4,4-bis(4-hydroxyphenyl)heptane, 2,2-(3,5-3',5'-tetrachloro-4,4'-dihydroxydiphenyl)propane, 2,2-(3,5,3',5'-tetrabromo-4,4'-dihydroxydiphenyl)propane, (3,3'-dichloro-4,4'-dihydroxyphenyl)methane. Other halogenated and non-halogenated diphenols of the bisphenol type can also be used such as are disclosed in U.S. Patents 2,999,835, 3,028,365 and 3,334,154.

It is possible to employ two or more different diphenols or a copolymer with a glycol or with hydroxy or acid terminated polyester, or with a dibasic acid in the event a carbonate copolymer or interpolymer rather than a homopolymer is desired for use in preparing the aromatic polycarbonate. Blends of any of these materials can also be used to obtain the aromatic polycarbonates.

These diphenols can then be employed to obtain the high molecular weight aromatic polycarbonates of the invention which can be linear or branched homopolymers or copolymers as well as mixtures thereof or polymeric blends and which generally have an intrinsic viscosity (IV) of about 0.40-1.0 dl/g as measured in methylene chloride at 25°C.



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The carbonate precursor used can be either a carbonyl halide, a carbonate ester or a haloformate. The carbonyl halides can be carbonyl bromide, carbonyl chloride and mixtures thereof. The carbonate esters can be diphenyl carbonate, di-(halophenyl) carbonates such as di-(chlorophenyl) carbonate, di-(bromophenyl) carbonate, di-(trichlorophenyl) carbonate, di-(tribromophenyl) carbonate, etc. di-(alkylphenyl) carbonate such as di(tolyl) carbonate, etc., di-(naphthyl) carbonate, di-(chloronaphthyl) carbonate, phenyl tolyl carbonate, chlorophenyl chloronaphthyl carbonate, etc., or mixtures thereof. The haloformates that can be used include bis-haloformates of dihydric phenols (bischloroformates of hydroquinone, etc.) or glycols (bishaloformates of ethylene glycol neopentyl glycol, polyethylene glycol, etc.). While other carbonate precursors will occur to those skilled in the art, carbonyl chloride, also known as phosgene, is preferred.

Also included are the polymeric derivatives of a dihydric phenol, a dicarboxylic acid and carbonic acid such as are disclosed in U.S. Patent 3,169,121 which is incorporated herein by reference, and which are particularly preferred. This class of compounds is generally referred to as copoly-estercarbonates.



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Molecular weight regulators, acid acceptors and catalysts can also be used in obtaining the aromatic polycarbonates of this invention. The useful molecular weight regulators include monohydric phenols such as phenol, chroman-I, paratertiarybutylphenol, parabromophenol, primary and secondary amines, etc. Preferably, phenol is employed as the molecular weight regulator.

A suitable acid acceptor can be either an organic or an inorganic acid acceptor. A suitable organic acid acceptor is a tertiary amine such as pyridine, triethylamine, dimethylaniline, tributylamine, etc. The inorganic acid acceptor can be either a hydroxide, a carbonate, a bicarbonate, or a phosphate of an alkali or alkaline earth metal.



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The catalysts which can be employed are those that typically aid the polymerization of the diphenol with phosgene. Suitable catalysts include tertiary amines such as triethylamine, tripropylamine, N,N-dimethylaniline, quaternary ammonium compounds such as, for example, tetraethylammonium bromide, cetyl triethyl ammonium bromide, tetra-n-heptylammonium iodide, tetra-n-propyl ammonium bromide, tetramethylammonium chloride, tetramethyl ammonium hydroxide, tetra-n-butyl ammonium iodide, benzyltrimethyl ammonium chloride and quaternary phosphonium compounds such as, for example, n-butyltriphenyl phosphonium bromide and methyltriphenyl phosphonium bromide.

Also included herein are branched polycarbonates wherein a polyfunctional aromatic compound is reacted with the diphenol and carbonate precursor to provide a thermoplastic randomly branched polycarbonate. These polyfunctional aromatic compounds contain at least three functional groups which are carboxyl, carboxylic anhydride, haloformyl, or mixtures thereof. Illustrative of polyfunctional aromatic compounds which can be employed include trimellitic anhydride, trimellitic acid, trimellityl trichloride, 4-chloroformyl phthalic anhydride, pyromellitic acid, pyromellitic dianhydride, mellitic acid, mellitic anhydride, trimesic acid, benzophenonetetracarboxylic acid, benzophenonetetracarboxylic anhydride, and the like. The preferred polyfunctional aromatic compounds are trimellitic anhydride and trimellitic acid or their acid halide derivatives.

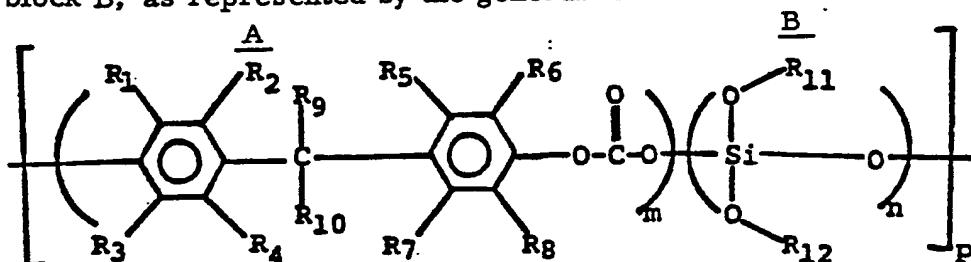


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Blends of linear and branched aromatic polycarbonates are also included within the scope of this invention.

Other well known materials can also be employed for their intended function and include such materials as anti-static agents, mold release agents, thermal stabilizers, ultraviolet light stabilizers, reinforcing fillers such as glass and other inert fillers, foaming agents, and the like.

The block copolymers that can be employed in the practice of this invention can be prepared by methods known to those skilled in the art, such as are disclosed in U.S. Patents 3,189,622 and 3,189,634 which are incorporated herein by reference. These block copolymers typically comprise alternating segments of polycarbonate and polyorganosiloxane; i.e., block A and block B, as represented by the general formula



wherein $R_1 - R_8$ can each be independently selected from the group consisting of hydrogen, halogen, alkyl having 1 to 6 carbon atoms and aryl; R_9 and R_{10} can each be independently selected from the group consisting of hydrogen, alkyl having 1 to 6 carbon atoms and aryl; R_{11} and R_{12} can each be independently selected from the group consisting of alkyl having 1 to 6 carbon atoms and aryl wherein the aryl can also form a ring member; m is an integer of about 1-10; n is an integer of about 5-100; and, p has a value of at least 1.



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Preferably, the polycarbonate segment of the block copolymer is derived from the same diphenol homopolymer as is the polycarbonate resin with which the block copolymer is to be blended. For example, if the polycarbonate resin is derived from the diphenol, bisphenol-A; i. e., BPA(2, 2-bis (4-hydroxphenyl)propane), then the polycarbonate segment of the block copolymer is preferably derived from the same diphenol; i. e., BPA. Or, if the polycarbonate resin is derived from the diphenol 2, 2-bis(4-hydroxy-3-methylphenyl) propane, then the polycarbonate segment of the block copolymer is derived from the same diphenol; i. e., 2, 2-bis(4-hydroxy-3-methylphenyl)propane, and so forth.

The polyorganosiloxane segment of the block copolymer is preferably polydimethylsiloxane (PDMS).

Preferred Embodiment of the Invention

The following examples are set forth to more fully and clearly illustrate the present invention and are intended to be, and should be construed as being, exemplary and not limitative of the invention. Unless otherwise stated, all parts and percentages are by weight.

Example 1

One hundred (100) parts of an aromatic polycarbonate was prepared by reacting BPA (2, 2-bis(4-hydroxyphenyl) propane) and phosgene in the presence of an acid acceptor and a molecular weight regulator. The resultant high molecular weight aromatic polycarbonate had an intrinsic viscosity (IV) of 0.50. This aromatic polycarbonate was subsequently mixed with the various block copolymers described in the ensuing examples by tumbling the ingredients together in a



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laboratory tumbler. In each instance, the resulting mixture was then fed through an extruder which was operated at about 285°C and the extrudate was comminuted into pellets.

The pellets were then injection molded at about 315°C into test bars of about (5 in.) 12.7cm by $\frac{1}{2}$ in. by about (1/16-1/8 in.) 0.16 -0.32cm thick and into test squares of about 5x5cm (2 in. by 2 in.) by about (1/8 in.) 0.32cm thick.

Example 2

A block copolymer consisting of a polycarbonate segment derived from BPA and polydimethylsiloxane (PDMS) in the polyorganosiloxane segment was prepared in accordance with the method disclosed in U.S. Patent 3,189,622. That is the block copolymer was prepared by forming a mixture of BPA and PDMS at a temperature of about 25°-100°C in the presence of an acid acceptor and phosgenating the mixture until the mass achieved a maximum viscosity. The resultant block copolymer consisted of 50% by weight polycarbonate segments and 50% by weight PDMS segments.

Example 3-6

The block copolymer of Example 2 was mixed with the aromatic polycarbonate of Example 1 at the weight percentages shown below and each of the mixtures was then extruded into pellets which were then molded into test bars and test squares following the procedure described in Example 1.



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	<u>Example 2</u>	<u>Example 1</u>
<u>Example</u>	<u>Block Copolymer</u>	<u>Aromatic Polycarbonate</u>
<u>Example</u>	<u>(Wt. %)</u>	<u>(Wt. %)</u>
3	1	99
4	3	97
5	5	95
5	10	90

Example 7

Following the procedure of Example 2, a block copolymer was obtained consisting of 35% by weight 10 polycarbonate segments and 65% by weight PDMS segments.

Example 8

Following the procedure of Example 1, 5% by weight of the block copolymer of Example 7 was mixed 15 with 95% by weight of the aromatic polycarbonate of Example 1 whereupon the mixture was extruded into pellets and the pellets molded into test bars and test squares as described in Example 1.

Example 9

20 The procedure of Example 2 was used to prepare a block copolymer consisting of 95% by weight polycarbonate segments and 5% by weight PDMS segments. This block copolymer was then extruded into pellets and the pellets molded into test bars and test 25 squares as described in Example 1.



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Example 10

A mixture of 97% by weight of the polycarbonate of Example 1 and 3% by weight PDMS was prepared, which was then extruded into pellets and the pellets molded into test bars and test squares following the procedure 5 of Example 1. In contrast to the smooth, homogeneous appearance of test bars and test squares obtained from mixtures of the aromatic polycarbonate of Example 1 with the block copolymers of Examples 3-6 and 8, the test bars and test squares obtained 10 from the aromatic polycarbonate-PDMS mixture of this example had a mottled, laminar appearance which could not be used as a commercially acceptable product.

The test bars and test squares of Examples 1, 3-6 15 and 8-10 were subject to various tests to determine various properties of the compositions. The test results wherein 5 test bars and 5 test squares were used for each test are set forth in Tables I and II below wherein the various tests were determined in 20 accordance with the following methods:

Flame retardancy was determined according to Underwriters' Laboratories, Inc. Bulletin UL-94, Burning Test for Classifying Materials. In accordance with this test procedure, materials so 25 investigated are rated either V-0, V-I or V-II based on the results of 5 specimens. The criteria for each V (for vertical) rating per UL-94 is briefly as follows:



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"V-0": Average flaming and/or glowing after removal of the igniting flame shall not exceed 5 seconds and none of the specimens shall drip flaming particles which ignite absorbent cotton.

5

"V-I": Average flaming and/or glowing after removal of the igniting flame shall not exceed 25 seconds and the glowing does not travel vertically for more than 1/8" of the specimen after flaming ceases and glowing is incapable of igniting absorbent cotton.

10

"V-II": Average flame and or glowing after removal of the igniting flame shall not exceed 25 seconds and the specimens drip flaming particles which ignite absorbent cotton.

15

In addition, a test bar which continues to burn for more than 25 seconds after removal of the igniting flame is classified, not by UL-94, but by the standards of the instant invention, as "burns".

Flexural modulus was determined in accordance with ASTM D-790; flexural yield was determined in accordance with ASTM D-790; unnotched and notched Izod impact strengths were determined in accordance with ASTM D-256; flammability oxygen ratio (Fenimore/Martin) was determined in accordance with ASTM D-2863; solvent resistance was evaluated by measuring the percent strain necessary to cause crazing in test samples exposed to one drop of solvent for a period of 3 minutes; and RDT (Retention of Ductility Time) denotes the maximum number of hours for which a test bar can be aged at



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a temperature before the mode of failure in the notched Izod impact test (ASTM-256) changes from ductile to brittle. Unless otherwise specified, the RDT refers to heat aging at 125°C of test bars

5 1/8" thick.

In Table I, "Skydrol" identifies a commercially obtained hydraulic fluid particularly deleterious to polycarbonates.



TABLE I
Physical Properties of Polycarbonate Blends

<u>Example</u>	<u>Block Copolymer Additive</u>	<u>Kg/cm² x 10⁻⁴</u>	<u>Flexural Modulus (psi x 10⁻⁵)</u>	<u>Yield Strength (psi x 10⁻³)</u>	<u>Unnotched Izod (ft.lbs./in., Kg - cm/cm)</u>	<u>Notched Izod (ft.lbs./in., Kg - cm/cm)</u>
			<u>Kg/cm² x 10⁻²</u>	<u>Kg/cm² x 10⁻²</u>	<u>(ft.lbs./in., Kg - cm/cm)</u>	<u>(ft.lbs./in., Kg - cm/cm)</u>
1	-	(3.32)	2.33	(14.1)	9.91	(>40) > 220
3	1.0	(3.42)	2.40	(13.5)	9.49	(>40) > 220
4	3.0	(3.26)	2.29	(13.1)	9.21	(>40) > 220
5	5.0	(3.24)	2.28	(12.9)	9.07	(>40) > 220
6	10.0	(2.3)	1.6	(12.4)	8.72	(>40) > 220
8	5.0	(3.22)	2.26	(12.5)	8.79	(>40) > 220
9	10.0	(2.89)	2.03	(11.6)	8.16	(>40) > 220
*10	3.0	(3.18)	2.24	(13.3)	9.35	(>40) > 220

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* PDMS used in place of block copolymer



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TABLE I (continued)
Physical Properties of Polycarbonate Blends

<u>Example</u>	<u>Solvent Resistance (% Strain)</u>		<u>Flammability (Fenimore/Martin)</u>		<u>UL-94 Rating</u>
	<u>Skydrol</u>	<u>Gasoline</u>	<u>CCl₄</u>	<u>O₂ Ratio</u>	
1	0.7	0.3	0.4	0.26	Burns
3	0.6	0.3	-	-	-
4	0.7	0.4	-	-	-
5	1.0	0.5	0.7	-	V-II
6	5.0	0.6	1.0	0.3	Burns
8	1.0	0.5	-	-	V-II
9	0.8	0.3	-	0.32	V-I
*10	-	-	-	>0.28	

*PDMS used in place of block copolymer



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TABLE II
Notched Izod Impact Strength (ft.-lbs./in.) kg-cm/cm
vs

<u>Example</u>	<u>Time at 125°C</u>				<u>RDT</u> 6 hrs.
	<u>6 Hrs.</u>	<u>7 Hrs.</u>	<u>16 Hrs.</u>	<u>90 Hrs.</u>	
1 (3.3) 18	(2.4)	14	(2.5)	14	(1.6) 8.7 6 hrs.
3 (8.2) 45	(2.7)	15	(2.8)	15	(1.7) 9.2 6 hrs.
4 -	-	(15.8)	86	(14.1) 77	(5.0) 27 2 wks.
5 -	-	-	-	(15.1) 82	(14.3) 77.8 > 2 wks.
6 (14.4) 78	-	(15.1)	82	(13.6) 74	(13.5) 73.4 > 2 wks.
8 -	-	-	-	(14.1) 77	(14.4) 78 > 2 wks.
9 -	-	-	-	(11.4) 62	(4.5) 24 2 wks.
10 (13.6) 74	-	(14.3)	78	(12.6) 69	(11.3) 61 > 2 wks.



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In Table I, it can be seen that as little as 3% of the Example 4 block copolymer was sufficient to improve solvent resistance against Skydrol and that 5% of the block copolymer of Example 5 raised the 5 resistance to 1% strain, which is about the maximum generally encountered in most practical situations.

Table I also reveals that, at higher levels of block copolymer, oxygen index and UL-94 ratings are improved (for instance Example 1 vs. Example 6).

From Table II it can be seen that use of the 10 block copolymers of the invention results in a larger positive effect on ductility and a smaller negative effect on molded properties of the test samples as shown by Examples 3-6 and 8. For example, 15 as little as 3% of the block copolymer was sufficient to extend the retention ductile impact behavior during aging at 125°C from 6 hours to at least 2 weeks (Example 4) and as little as 5% of the block copolymer was sufficient to eliminate any brittle 20 impact behavior after aging for two weeks at 125°C (Examples 5 and 8).

The improvement obtained by mixing the block copolymer of the invention with the aromatic polycarbonate as opposed to using the block copolymer alone can be readily seen by comparing the results 25 of Example 6 with Example 9 in Table II. Although each example contains 5% PDMS, Example 6 consisting of the aromatic polycarbonate-block copolymer mixture retained its impact strength and 30 ductility after two weeks of aging at 125°C whereas Example 9, consisting of only the block copolymer, did not.



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Example 11

The procedure of Example 1 was repeated except that 0.5% by weight of the sodium salt of trichlorobenzene sulfonic acid (STB) and 0.1% by weight polytetrafluoroethylene (PTFE) were mixed 5 with the aromatic polycarbonate. The mixture was extruded into pellets as in Example 1, but instead of injection molding the pellets into test bars and test squares, the pellets were extruded into sheets measuring 4 feet square by 0.318 cm (0.125") 10 thick.

Example 12

The procedure of Example 2 was repeated to obtain a block copolymer consisting of 57% by weight polycarbonate segments and 43% by weight PDMS.

Example 13

The procedure of Example 11 was repeated except that 4% by weight of the block copolymer of Example 12 was mixed with the other ingredients of Example 11 to obtain the aromatic polycarbonate 20 sheets.

The sheets obtained from Examples II and 13 were then subjected to solvent exposure to determine the stress levels necessary to induce stress crazing during given time periods. The 25 results obtained are set forth in Tables III and IV below wherein "Spray Nine", "Lexsol" and "Royalite S-22" identify commercially obtained cleaning fluids.



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<u>Solvent</u>	<u>Stress Level After One Hour Exposure</u>	
	<u>Example II</u>	<u>Example I₃</u>
Carbon Tetrachloride	(500) 35	(1000) 70
Toluene	(500) 35	(1000) 70
Benzene	(500) 35	(1000) 70
Butyl Cellosolve	(1500) 105	(2000) 70
Isopropyl Alcohol	(>2500)>176	(>2500)>176
Methyl Alcohol	(500) 35	(1000) 70
Royalite S-22	(2000) 141	(>2500)>176
Spray Nine	(>2500)>176	(>2500)>176
Lexsol		



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TABLE IV

Time to Stress Crack at 1500 PSI Stress With
Continuous Wetting

<u>Solvent</u>	<u>Example 11</u>	<u>Time</u>	<u>Example 13</u>
Carbon Tetrachloride	1 min.		38 min.
Gasoline	immediate		~1 min.

5 The results in Tables III and IV above reveal
that Example 13, containing the additional 4% by
weight of the block copolymer required higher stress
levels to induce stress-crazing than did Example 11.
10 Example 13 was also more durable than Example 11
when exposed to carbon tetrachloride as shown in
Table IV. In general, the results in Tables III
and IV indicate that improved solvent resistance
is obtained when the aromatic polycarbonate is
further modified with the block copolymer.

15 Example 14
The procedure of Example 1 was followed to
prepare aromatic polycarbonate test bars and test
squares comprising 70% by weight of the polycarbonate
of Example 1 and 30% by weight of the block copolymer
20 of Example 12. The properties of Example 1 were
compared with those of this example (14) and the
results are set forth in Table V below wherein
tensile strength (psi), elongation (%), and modulus
(psi) results were determined in accordance with
25 ASTM D-638.



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TABLE V
Comparative Properties of Example 14 and Example 1

<u>Properties</u>	<u>Example 14</u>	<u>Example 1</u>
Tensile Strength (psi) kg/cm ²	(6700) 471	(9500) 670
Elongation (%)	155	110
Modulus (psi) kg/cm ²	10.5 (150) x 10 ³	24.2 (345) x 10 ³
UL-94 Rating	V-0	V-II
Time to Stress Crack @ 1500 psi	No crack	
with carbon tetrachloride	>1 hr.	<1 min.
with gasoline	>1 hr.	<1 min.
Stress level to crack after 1 1/2 hr. continuous exposure (psi) kg/cm ²		
with carbon tetrachloride	*No crack	77.3 (110)
with butyl cellosolve	*No crack	130-140 (1800-2000)
with Royalite C	*No crack	35 (500)

* Surface etch was noted after (500, 2500 and 5000) psi, respectively, but test bars remained completely ductile upon bending.



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The results in Table V above reveal the dramatic improvement in solvent resistance obtained with the aromatic polycarbonate-block copolymer mixture (Example 14) as opposed to the unmodified aromatic 5 polycarbonate (Example 1).

Examples 15-17

The block copolymer of Example 12 was mixed with the aromatic polycarbonate of Example 1 and the 10 aromatic polycarbonate-PDMS mixture of Example 11 at the weight percentages shown below and test sheets of these mixtures were prepared as described in Example 11.



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Example 15-17 (continued)

<u>Example</u>	<u>Aromatic Polycarbonate (Wt. %)</u>	<u>Example 10 Aromatic Polycarbonate- STB/PTFE Mixture (Wt. %) STB</u>	<u>Example 12 Block Copolymer (Wt. %)</u>
15	92.5	-	7.5
16	96.0	0.5	0.1
17	76.0	0.5	0.1
			30.0



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These Examples were subjected to various tests, the results of which are set forth in Table VI below wherein tensile yield strength (psi) and ultimate strength (psi) were determined in accordance with ASTM D-638.



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TABLE VI
Comparative Properties of Test Bars and Test Sheets

<u>Properties</u>	<u>Examples</u>		
	<u>1</u>	<u>14</u>	<u>15</u>
<u>Tensile:</u>			
Yield Strength (psi)kg/cm ²	(9000) 630	-	(9000) 630
Ultimate Strength (psi)kg/cm ²	(9500) 670	(6800) 480	(11520) 810
Elongation (%)	110	155	110
Modulus (psi)kg/cm ² x 10 ³	(345) 24.3	(150) 10.5	-
Flexural Modulus (psi)kg/cm ²	(340) 23.9	-	(250) 17.6
<u>Solvent Resistance:</u>			
Time to crack at (1500 psi) 105 kg/cm ²	> 60	> 60	> 60
• with carbon tetrachloride (min.)	~ 1	< 1	< 1
• with gasoline (min.)			25
Stress to crack after 1 hr. exposure (psi)			
• with carbon tetrachloride	(1100) 77	> 2500	176
• with butyl cellosolve	(1900) 135	(2500)	176
• with Royalite S-22	(500) 35	(2500)	176
• with gasoline	(1000) 70	-	(2500) 175



TABLE VI (continued)
Comparative Properties of Test Bars and Test Sheets

<u>Properties</u>	<u>Examples</u>		
	<u>11</u>	<u>13</u>	<u>17</u>
<u>Tensile:</u>			
Yield Strength (psi) kg/cm ²	(9000) 630	(9260) 650	(9200) 650
Ultimate Strength (psi) kg/cm ²	(8500) 600	(8500) 600	(9000) 630
Elongation (%)	95	70	85
Modulus (psi) kg/cm ² x 10 ³	(325) 22.8	-	-
Flexural Modulus (psi) kg/cm ²	(325) 22.8	(265) 18.6	(250) 17.6
Solvent Resistance:			
Time to crack at (1500 psi) 105 kg/cm ²			
with carbon tetrachloride (min.)	~1	38	>60
with gasoline (min.)	<1	1	>60
Stress to crack after 1 hr. exposure (psi)			
with carbon tetrachloride	(<500) <35	(1000) 70	(2500) 175
with butyl cellosolve	(1500) 105	(2000) 140	(1500) 105
with Royalite S-22	(500) 35	(1000) 70	(2000) 140
with gasoline	(<500) <35	(<500) <35	(1000) 70

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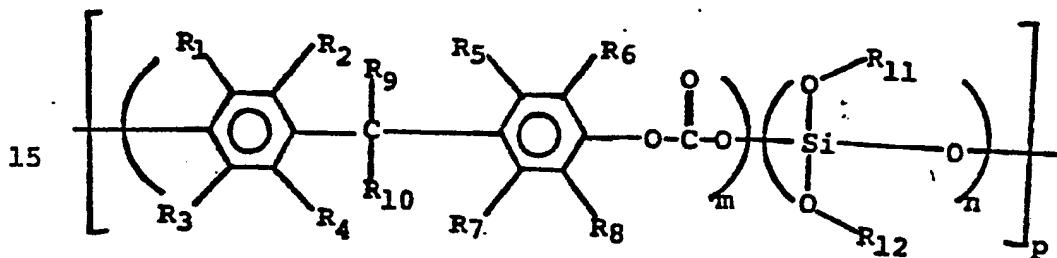
The results shown in Table VI above reveal that while the tensile properties of the block copolymer of the invention (Examples 13-17) compared favorably with the aromatic polycarbonate without the block copolymer additive (Examples I and II), the solvent resistance and stress crack resistance were notably better.



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Claims

1. A ductile, solvent resistant aromatic polycarbonate composition having improved flame retardance, said composition comprising a mixture of a high molecular weight aromatic polycarbonate and a block copolymer in an amount of about up to 50% by weight of said aromatic polycarbonate, said block copolymer having alternating segments of aromatic polycarbonate and polyorganosiloxane represented by the general formula

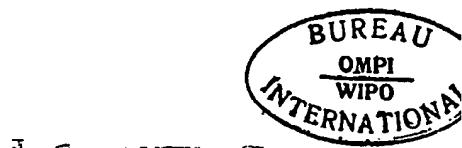


wherein R₁-R₈ can each independently be selected from the group consisting of hydrogen, halogen alkyl having 1 to 6 carbon atoms, and aryl; R₉ and R₁₀ can each be independently selected from the group consisting of hydrogen, alkyl having 1 to 6 carbon atoms, and aryl; R₁₁ and R₁₂ can each be independently selected from the group consisting of alkyl having 1 to 6 carbon atoms, and aryl; m is an integer of about 1-10; n is an integer of about 5-100; and, p has a value of at least 1, the weight ratio of said polycarbonate segments to said polyorganosiloxane segments in said block copolymer being in the range of about 25:75-75:25.



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2. The composition of claim 1 wherein the aromatic polycarbonate and the polycarbonate segments in said block copolymer are each derived from the same diphenol homopolymer.
3. The composition of claim 2 wherein the diphenol homopolymer is 2,2-bis(4-hydroxyphenyl)propane.
4. The composition of claim 1 wherein the polyorganosiloxane segments are polydimethylsiloxane.
5. The composition of claim 1 which includes about 0.1-1.0 weight percent of the sodium salt of trichlorobenzene sulfonic acid and about 0.05-0.3 weight percent polytetrafluoroethylene.



INTERNATIONAL SEARCH REPORT

International Application No. PCT/US79/00428

WO 80/00084

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) ¹

According to International Patent Classification (IPC) or to both National Classification and IPC
 INT. CL. C08L 83/10; 69/00
 U.S. CL. 525/464; 525/474

II. FIELDS SEARCHED

Minimum Documentation Searched ⁴

Classification System	Classification Symbols
US	525/464 525/474

Documentation Searched other than Minimum Documentation
to the Extent that such Documents are Included in the Fields Searched ⁵

III. DOCUMENTS CONSIDERED TO BE RELEVANT ¹⁶

Category ¹⁶	Citation of Document, ¹⁶ with indication, where appropriate, of the relevant passages ¹⁷	Relevant to Claim No. ¹⁶
X	US, A 3,686,355 PUBLISHED 22 AUGUST 1972 GAINES ET AL see column 5, lines 15-23, column 6, lines 7-12 and sample N ^c of Table II	1-5
A	US, A, 3,742,085 PUBLISHED 26 JUNE 1973 BIALOUS	5
A	US, A, 4,073,768 PUBLISHED 14 FEBRUARY 1978 MARK	5

* Special categories of cited documents: ¹⁵

"A" document defining the general state of the art

"E" earlier document but published on or after the international filing date

"L" document cited for special reason other than those referred to in the other categories

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but on or after the priority date claimed

"T" later document published on or after the international filing date or priority date and not in conflict with the application, but cited to understand the principle or theory underlying the invention

"X" document of particular relevance

IV. CERTIFICATION

Date of the Actual Completion of the International Search ¹⁸

21 SEPTEMBER 1979

Date of Mailing of this International Search Report ¹⁸

18 OCT 1979

International Searching Authority ¹⁹

ISA/US

Signature of Authorized Officer ²⁰

W. J. BRIGGS